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Conformation and Dynamics of Polymers in Solution

Final Report

Ben Chu

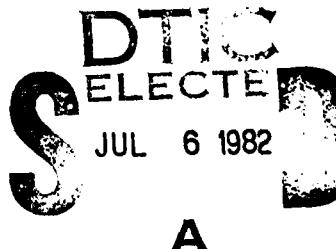
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Polymer Dynamics, Phase Transitions, Polymer Kinetics, Light Scattering.		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number)		

Foreward

I have been supported by the U.S. Army Research Office without interruption since 1963. The financial assistance and the long range goals of the program administrators including their patience and understanding are greatly appreciated. I have been working towards the improvements in instrumentation related to laser light scattering and its applications to polymer physics and am glad to report that the field has finally reached a stage capable of contributing towards the basic as well as applied aspects of polymer science.



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Final Report

a. Statement of problems studied.

- (i) To investigate the static and dynamical properties of polymers in solution from dilute to semidilute solutions using the scaling concept.
- (ii) To examine the characterization of molecular polydispersity effects using photon correlation function analysis.
- (iii) To study the temperature-composition behavior of a polymer solution, polystyrene in methyl acetate, which has an upper and a lower critical solution temperature.
- (iv) To study polymerization kinetics by Raman scattering.

b. Summary of the most important results.

- (i) We have shown that although the scaling concept is correct in the limit of infinite polymer molecular weight, its application needs to be modified for finite molecular weight polymers. In semidilute solution, the presence of pseudo gel high frequency motions can be analyzed by means of a histogram method of correlation function profile analysis.
- (ii) In our characterization of molecular polydispersity, we must emphasize the interactive nature of experimental measurements, methods of data analysis, and comparison with theory. An empirical method was developed to approximate the Laplace inversion in order to estimate the linewidth distribution function. By taking into account the particle scattering factor and intermolecular interactions, we were able to determine the molecular weight distributions of polymers such as polystyrene, poly[bis(m-chlorophenoxy) phosphazene].
- (iii) We have examined a polymer solution, polystyrene in methyl acetate PS/MA which has an upper and a lower critical solution temperature within the

experimental range of our light scattering spectrometer. The result of this investigation has led us to consider the PS/MA system as a possible candidate for studying the kinetics of spinodal decomposition and nucleation processes.

(iv) Raman scattering in combination with light scattering and refractive index measurements may permit us to develop a technique to study polymerization kinetics including the determination of polymer/monomer concentration, molecular weight and distribution of polymer, as well as polymer size. (See attached reprint and galley).

c. List of publications.

During the contract period (4/1/79-3/31/82) the following articles were published with the support of the U.S. Army Research Office.

102. Esin Gulari, Erdogan Gulari, Y. Tsunashima and B. Chu, Photon Correlation Spectroscopy of Particle Distributions, J. Chem. Phys., 70(8), 3965 (1979).
103. B. Chu, Dynamics of Macromolecular Solutions, Physica Scripta, 19, 458 (1979).
104. B. Chu, Esin Gulari and Erdogan Gulari, Photon Correlation Measurements of Colloidal Size Distributions. II. Details of Histogram Approach and Comparison of Methods of Data Analysis, Physica Scripta, 19, 476 (1979).
- 105.* B. Chu and T. Nose, Static and Dynamical Properties of Polystyrene in Transdecalin, Macromolecules, 12, 347 (1979).
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 129. Y.-H. Lin, G. Fytas and B. Chu, Depolarized Rayleigh Spectra of Siloxane Polymers, J. Chem. Phys. 75, 2091 (1981).

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Study of Thermal Polymerization of Styrene by Raman Scattering

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Department of Chemistry, State University of New York at Stony Brook,
Long Island, New York 11794. Received May 22, 1980

ABSTRACT: We present a new method for studying polymerization kinetics based on Raman scattering. Our method has selectivity, is reliable and easy to apply, can be automated, and is insensitive to most spurious phenomena. We demonstrate this method by a study of the thermal polymerization of styrene at 60, 75, and 90 °C.

Introduction

The thermal polymerization of styrene has recently been the subject of several studies by light-beating spectroscopy (polarized¹ and depolarized²), interferometry (polarized³ and depolarized⁴), and NMR.⁵ We present here a new method based on Raman scattering. Our method has selectivity, is reliable and easy to apply, can be automated, and involves little external calibration. We emphasize that, since polymerization involves changes in chemical bonds which can usually be observed through Raman lines, this technique should be applicable to almost any polymerization process.

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Principle and Calibration

The principle of the method is to record Raman spectra from the sample at different stages of polymerization. In a polymerization reaction some portion of the molecular structure of interest usually remains unchanged. The corresponding Raman-active lines are present throughout the process and can be used as standards to calibrate the intensity of other lines, while those related specifically to the monomer vanish and those related to the polymer grow. Monitoring a ratio of intensities makes the measurement insensitive to laser power fluctuations or sample turbidity. Therefore, the method is reliable even in unfavorable conditions. In particular, applications of the method for studying copolymerization processes should be a worthwhile extension.

The first step is to measure Raman spectra of the monomer and of the polymer and to select the useful lines. When many Raman peaks are present, some of this in-

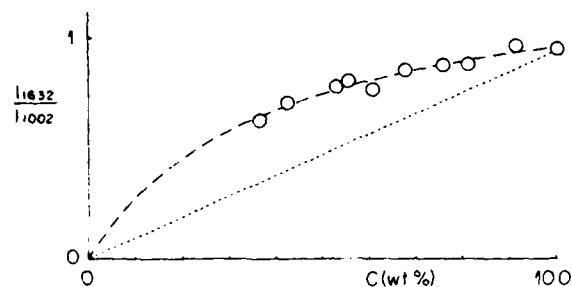


Figure 1. Ratio of the intensities of the 1632-cm^{-1} (double bond) line to the 1002-cm^{-1} (ring) line, I_2/I_1 , as a function of the monomer concentration: (---) equal-scattering hypothesis; (—) fit to eq 2.

formation is redundant. Consequently, there is no need to scan the entire Raman spectrum during kinetic studies. Furthermore, for many interesting polymers the spectra and assignments of both the monomers and the polymers are available in the literature. In our case we selected the 1632-cm^{-1} line due to the aliphatic double bond (denoted by subscript 2) and the 1002-cm^{-1} line due to a ring vibration (denoted by subscript 1) for our studies of thermal polymerization of styrene. A natural hypothesis is to consider the ring lines independent of the concentration. In other words, the Raman scattering due to the ring vibration remains the same whether it belongs to a monomer or to a polymer. Unfortunately, this is misleading. Indeed we measured the ratio of those lines in polystyrene in styrene solutions of different known polymer concentrations. The above assumption predicts straight-line behavior, obviously not verified by the data, as shown in Figure 1.

Let c_m be the concentration of monomer molecules and c_p be that of polymer segments in a unit volume. The intensity due to the aliphatic double bond is $I_2 = a_2 c_m$ and that due to the ring is $I_1 = a_1 c_m + b_1 c_p$, where $a_1 \neq b_1$. So we get

$$\frac{I_2}{I_1} = \frac{a_2 c_m}{a_1 c_m + b_1 c_p} \quad (1)$$

If the concentrations appear only as ratios, we can use mass concentrations and normalize them so that $c_m + c_p = 1$. Then we have the empirical expression

$$\frac{I_2}{I_1} = \frac{a_2}{a_1} c_m \left[\frac{1}{b_1/a_1 + (1 - b_1/a_1)c_m} \right] \quad (2)$$

The equations are insensitive to density changes. The ratio a_2/a_1 is equal to I_2/I_1 when $c_m = 1$. The ratio b_1/a_1 can be deduced from a fit of the calibration data in Figure 1. The upper curve in Figure 1 represents a good agreement between eq 2 and our calibration using $a_2/a_1 = 1.32 \pm 0.06$ and $b_1/a_1 = 0.59 \pm 0.04$. The first of these ratios has a weak temperature dependence but the second, corresponding to lines at the same frequency, can be assumed constant and is essentially independent of the optical alignment of the Raman spectrometer. For more complex systems, other empirical formulas can be developed. In any case, a calibration is essential in order to achieve quantitative results.

Thermal Polymerization of Styrene

We have applied this technique to the thermal polymerization of styrene, using a Spex 1302 double-monochromator (0.5 m, 1200 grooves/mm) and recording the spectra by means of a chart recorder. As we were interested mainly in the integrated intensities of the peaks, we could use wide slits ($\sim 300\text{ }\mu\text{m}$), which reduce the required laser power

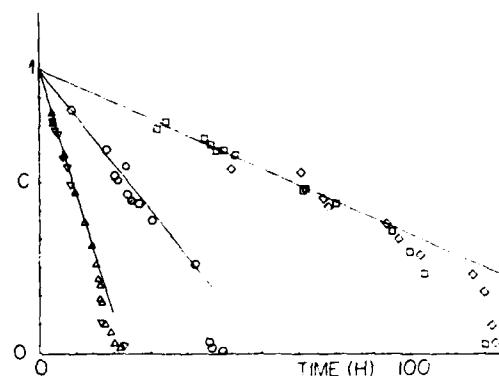


Figure 2. Concentration of monomer vs. time: (▼, △) cells 1 and 4, 90 °C; (○, ○) cells 2 and 5, 75 °C; (□, □) cells 3 and 6, 60 °C. H = hour.

Table I

	temperature, °C		
	60	75	90
initial rate, %/h	0.32	1.1	3.1

($\sim 300\text{ mW}$ at 488 nm) and thence the occurrence of any photoactivated process. In fact, a spectrometer of much poorer quality would have been sufficient. On the other hand, refinements such as computer-controlled scan, digital recording, and automatic data acquisition can be introduced easily. We are in the process of making our experiments fully automatic.

Among the six samples we have studied, five were only degassed by several freeze-pump cycles, the inhibitor (*tert*-butylpyrocatechol) being left undisturbed. The sixth one was washed several times with water in order to remove the inhibitor and then dried, first with calcium chloride and then with calcium hydride. The observed rate of polymerization for that cell (no. 1) agrees quite well with the other measurements. We ran two samples at each of three temperatures, 60, 75, and 90 °C, and found good reproducibility in our measurements. The concentration of monomer vs. time is reported in Figure 2 for all cells. We find a linear behavior until about 65% of the reaction, where it suddenly becomes faster. Such a behavior, known as the Trammsdorf effect,⁶ has been observed by light scattering techniques.⁴ It is not clear yet whether this is a real phenomenon or an experimental artifact. Indeed when the viscosity of the sample becomes very high, the portion lighted is no longer renewed so the effect of several hundred milliwatts of light may no longer be negligible. In that region we have actually observed a thermal lens effect which has persisted hours after stopping the beam, but we cannot yet ascertain whether this effect is the reason for the change in the kinetics. Another possible reason for the acceleration in the polymerization kinetics is due to a decrease of the termination rate constant, in terms of intrachain interactions⁶ and chain-transfer effect,⁷ through tremendous viscosity increases during the polymerization process.

From the results of Figure 2, we can infer a variation of the initial rate with the temperature, as listed in Table I. The results are very well described by an Arrhenius law with an activation energy of 19 kcal/mol, the same as observed by classical methods,⁸ as can be seen in Figure 3. On the other hand, the rates we found are about 3 times higher than usually reported. We believe this disagreement is possibly due to sample preparation procedures and not the observation method. In order to check the Raman detection method, we polymerized an additional styrene

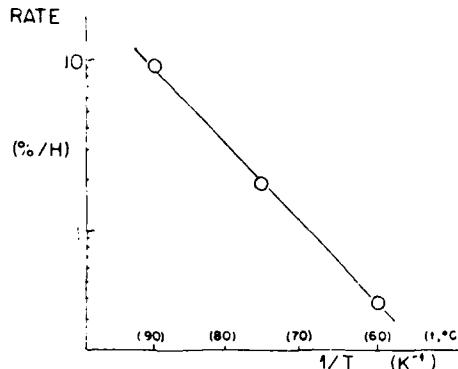


Figure 3. log of the initial reaction rate vs. reciprocal temperature. The solid line represents an Arrhenius law with an activation energy of 19 kcal/mol. H = hour.

sample at 75 °C for 19 h, during which time we monitored the monomer concentration, as shown typically in Figures 1 and 2. After stopping the reaction, we determined the polymer conversion classically by first dissolving the polymerized sample in 1,4-dioxane and then precipitating the polystyrene in methyl alcohol.⁸ We found an 18% conversion by the classical method and a 21% conversion based on Raman scattering. The agreement is quite reasonable, especially in view of the fact that there must be a residual amount of oligomers which are not precipitated by the classical method.

Conclusion

We have presented a new method for studying polym-

erization reactions based on Raman scattering. This method is fairly general, easy to use and to automate, and insensitive to most spurious phenomena. We have illustrated this method by a study of the thermal polymerization of styrene at different temperatures and found our results to be in essential agreement with those obtained by other methods.

Acknowledgment. We gratefully acknowledge support from the U.S. Army Research Office. G.Z. gratefully acknowledges support from Conseiller Culturel de l'Am-bassade de France and the Service de l'Enseignement et des Echanges Linguistiques—Ministère des Affaires Etrangères (Paris). We thank Day-Chyuan Lee for his help in performing the Raman studies and the chemical analysis on the polystyrene content and Kirk J. Abbey for a helpful discussion.

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Light Scattering and Spectroscopic Studies of Polymerization Processes. 2. Thermal Polymerization of Styrene

Benedict Thomas Pugh-Wise Chair and Game Chair

Chemical Engineering Department, State University of New York at Stony Brook, Long Island, New York 11748, December 20, 1963

ABSTRACT: By means of thermal polymerization of styrene at temperatures up to 200°C., we have obtained a series of polymers which have been characterized by infrared, ultraviolet, and visible absorption spectra, viscometry, and electron spin resonance. The molecular weights of the polymers range from 10,000 to 100,000. The infrared spectra show absorption bands due to aromatic groups, aliphatic groups, and carbonyl groups. The ultraviolet spectra show absorption bands due to aromatic groups, aliphatic groups, and carbonyl groups. The visible spectra show absorption bands due to aromatic groups, aliphatic groups, and carbonyl groups.

1. Introduction

Studies of light scattering by polymers have been difficult because of the large number of factors which can influence the intensity of the scattered light. In this paper we shall discuss the effect of concentration, temperature, and other physical variables on the intensity of the scattered light. We shall also discuss the effect of the chemical structure of the polymer on the intensity of the scattered light.

Light scattering intensity measurements can be used to determine the molecular weight, the second virial coefficient, and the radius of gyration of a polymer. In this paper we shall discuss the effect of concentration, temperature, and other physical variables on the intensity of the scattered light. We shall also discuss the effect of the chemical structure of the polymer on the intensity of the scattered light.

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We shall also discuss the effect of concentration, temperature, and other physical variables on the intensity of the scattered light. We shall also discuss the effect of the chemical structure of the polymer on the intensity of the scattered light.

2. Experimental Methods

a. Light Scattering

Light scattering intensity measurements can be used to determine the molecular weight, the second virial coefficient, and the radius of gyration of a polymer.

The intensity of the scattered light is proportional to the square of the concentration of the polymer.

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concentrations of three chemicals. As our ultimate aim is to make and to construct a probe which can be used in a photometric system, we can use the above technique of measurement to determine the molecular weight of the polymer. In this article, we make use of Raman spectra for the precise determination of specific species, namely the reaction and for accurate measurements. Consequently, in the final probe design, a fairly low-resolution spectrometer for measuring the appropriate Raman peaks is likely to suffice.

b. Intensity of Scattered Light. In other subsections of this section, we have

$$\frac{HC}{R_0(A)} = \left(\frac{1}{M} - \frac{1}{R_0(A)} \right) + 2A_0 C + \dots \quad (1)$$

or $A(H) = (2A_0)^{-1} \ln \left(\frac{1}{M} - \frac{1}{R_0(A)} \right)$ is a constant dependent upon the concentration of the polymer, where M is the molecular weight, $R_0(A)$ is the concentration-dependent term, and A_0 and C are the molecular weight and the total virial coefficient. $R_0(A)$ is the excess Rayleigh ratio due to concentration fluctuations of the polymer solution using vertically polarized incident and scattered light and $A(H)$ is the particle scattering factor. Light scattering has been a standard technique for the determination of M , A_0 and the radius of gyration, r_g .

c. Absolute Scattered Intensity. Equation 1 is no longer valid at higher concentrations. Now we want to show that although we cannot yet derive a quantitative expression which will relate $HC(R_0(A))$ to the molecular weight at high concentrations, we obtain a unique and empirical relationship between $\ln \left(\frac{1}{M} - \frac{1}{R_0(A)} \right)$ and the molecular weight over a range of concentrations up to 10 molal. As one simple ex-

pression for the magnitude of the molecular weight at higher concentrations, we derive a quantitative expression which will relate $HC(R_0(A))$ to the molecular weight at high concentrations, we obtain a unique and empirical relationship between $\ln \left(\frac{1}{M} - \frac{1}{R_0(A)} \right)$ and the molecular weight over a range of concentrations up to 10 molal. As one simple ex-

$$\frac{HC}{R_0(A)} = \frac{(r_g/A)^2 r_p}{Rf} \quad (2)$$

where $K = (r_g/A) \ln \left(\frac{1}{M} - \frac{1}{R_0(A)} \right)$ is the magnitude of the non-magnetic dipole vector, the osmotic compressibility value of water has been used to relate to the value of the osmotic pressure.

b. Angular Distribution of Scattered Intensity

d. $E(X)$. The characteristic length X can be computed from the relation,

$$C(X) = A(X) / (1 + L(X)^2) \quad (3)$$

e. Finite dilution, $L = (r_g^2)^{1/2}$. In the small-angle region, L is related to a correlation length ζ ,

$$C(\zeta) = A(\zeta) + 4\pi(r_g^2)^{1/2} \quad (4)$$

where ζ is the clipping level, A is the background, and r_g is an unknown parameter in the chain-scattering procedure.

(r_g^2)^{1/2} is the decay time, τ , and Δ is the channel width and the instrument decay time, respectively, r_g^2/τ is the measured correlation function of the scattered light.

e. Translational Diffusion Coefficients?

f. A monodisperse system of spherically symmetric and non-interacting macromolecules in solution (or colloidal particles in suspension)

$$(A(t))^2 \ln \left(\frac{1}{M} - \frac{1}{R_0(A)} \right) = \frac{1}{M} K(t) \quad (5)$$

where the total scattered intensity $I(t) = I(M)$, with N

and t being the time of illumination in seconds (or colloidal particles in suspension).

D is the translational diffusion coefficient.

$A(t)$ is the diffusional amplitude.

$K(t)$ is the diffusional amplitude.

M is the molecular weight.

$R_0(A)$ is the concentration.

t is the time of illumination.

N is the number of molecules.

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polymerized in styrene (diluted in benzene). We find that the NBS 706 standard has a higher molecular weight than the NBS 705 standard. The only difference between the two standards is that our NBS sample seems to have a higher molecular weight even though it does not bear on our objectives. In view of the small molecular asymmetry observed for the NBS 706 standard polymer in styrene, we used a higher molecular weight polystyrene (polymer B, $M_w \sim 1.8 \times 10^7$ for dilution) for polymer B in styrene at 50 °C, where $R_{\text{c}} = 1.7 \times 10^7$. Therefore we have

$$R_{\text{c}}(M) = R_{\text{c}}(M_B) / (M_B/M)^2 \quad (10)$$

In Fig. 8, $R_{\text{c}} = 2.02 \pm 0.02$, $\sigma_r = 1$. From the slope of the linear plot of $R_{\text{c}}(M)$ versus M^{-1} we can determine the value of $R_{\text{c}}(M)$ at concentration $C = 25$ °C for each of our different molecular weight polymers A and B. From the initial slope and the intercept of the curve for polymer B we obtained $R_{\text{c}} = 1.7 \times 10^7$ cm² and $\sigma_r = 1.7 \pm 10^7$ g/mol. At higher concentrations, i.e., $(M/M_c) > 10$ larger errors in R_{c} are apparent as indicated by the schematic representation of Figure 9. Figure 10 shows the uppermost correlation of $R_{\text{c}}(M)$ versus M^{-1} for polymer A in a solvent. From Figure 10 the uppermost correlation appears to be at a lower concentration of polymer A in benzene. At a lower concentration of polymer A in benzene the same system turns out to be a fairly good approximation. The polymer end to some extent restricts the rotation.

In determining the osmotic compressibility ($\partial r/\partial C_{\text{P}}$) from scattering angle, we have to extrapolate the shoulder scattered intensity to zero scattering angle. We tried to construct a $(\partial r/\partial C_{\text{P}})^{-1} - C_{\text{P}}$ -plot and failed in order to construct the molecular scattering function $\chi(C_{\text{P}}, \theta)$ and r_{c} from the shoulder scattered intensity. This is due to the fact that the shoulder scattered intensity to zero scattering angle, i.e., we can construct a $R_{\text{c}}(10^7 \text{ C}_{\text{P}})$ surface as shown schematically in Figure 12. Again, the surface is not parallel to the $R_{\text{c}}(10^7 \text{ C}_{\text{P}})$ - $R_{\text{c}}(10^7 \text{ C}_{\text{P}})$ - $R_{\text{c}}(10^7 \text{ C}_{\text{P}})$ plane and the magnitude of the maximum R_{c} depends on the molecular weight. If we know R_{c} and C_{P} , we can find R_{c} at one point, whose coordinate on the $R_{\text{c}}(10^7 \text{ C}_{\text{P}})$ - $R_{\text{c}}(10^7 \text{ C}_{\text{P}})$ - $R_{\text{c}}(10^7 \text{ C}_{\text{P}})$ plane is $(R_{\text{c}}(10^7 \text{ C}_{\text{P}}), R_{\text{c}}(10^7 \text{ C}_{\text{P}}), R_{\text{c}}(10^7 \text{ C}_{\text{P}}))$. Then we can find R_{c} at another point, whose coordinate on the $R_{\text{c}}(10^7 \text{ C}_{\text{P}})$ - $R_{\text{c}}(10^7 \text{ C}_{\text{P}})$ - $R_{\text{c}}(10^7 \text{ C}_{\text{P}})$ plane is $(R_{\text{c}}(10^7 \text{ C}_{\text{P}}), R_{\text{c}}(10^7 \text{ C}_{\text{P}}), R_{\text{c}}(10^7 \text{ C}_{\text{P}}))$. This procedure can be repeated until we get the maximum R_{c} , although we have not yet worked out the most efficient ways of representing such a surface (or coordinate lines). In particular, while polystyrene is an easy system to work with and can be used as an illustration to construct the scheme, its practical value is more limited. Consequently, we are using that approach to study the non-symmetrical polymerization kinetics of benzene-chlorobenzene mixture and present them following in a later article.

B. Dynamic Properties. Figure 13 shows a typical plot of the net scattering function $A(C, \theta)$ as a function of many channel number i with $r = 4\pi$ for $C = 5.0 \times 10^7$ g/l, $\theta = 50^\circ$, $T = 25$ °C, and $d\tau = 8$ nm. By using the $A(C, \theta)$ as the experimental data, we obtained $\langle r^2 \rangle = 1.50 \times 10^7$ (cm²) and $\langle r^4 \rangle/(\langle r^2 \rangle)^2 = 0.13$. With a similar method, we can obtain the scattering function for the NBS 706 standard polymer in benzene solution. The results are shown in Figure 14. The variance for the NBS 706 standard polymer in benzene solution is $\langle r^2 \rangle = 1.50 \times 10^7$ (cm²) and $\langle r^4 \rangle/(\langle r^2 \rangle)^2 = 0.13$. Although the second-order cumulants for two different benzene solutions (70% and 100%) are almost the same, the percent deviation of Figures 13 and 14, the variance is distinctly different. Additional measurements of the NBS 706 standard in

FIG. 9 (067, 1-2)
FIG. 10 (068, 1-2)



FIG. 11 (090, 1-2), FIG. 12 (090, 1-2)

Σ_{UV}

FIG. 13 (090, 1-2)



of the net scattering function $A(C, \theta)$ as a function of many channel number i with $r = 4\pi$ for $C = 5.0 \times 10^7$ g/l, $\theta = 50^\circ$, $T = 25$ °C, and $d\tau = 8$ nm. By using the $A(C, \theta)$ as the experimental data, we obtained $\langle r^2 \rangle = 1.50 \times 10^7$ (cm²) and $\langle r^4 \rangle/(\langle r^2 \rangle)^2 = 0.13$. Although the second-order cumulants for two different benzene solutions (70% and 100%) are almost the same, the percent deviation of Figures 13 and 14, the variance is distinctly different. Additional measurements of the NBS 706 standard in

FIG. 14 (062, 1-2)
FIG. 15 (065, 1-2)

FIG. 16 (012, 1-2)
FIG. 17 (016, 1-2), FIG. 18 (018, 1-2)

FIG. 18 (018, 1-2)
FIG. 19 (018, 1-2)

where we have neglected the refractive index correction to the initial scattered intensity of the object molecule. Only the mean $\langle r^2 \rangle$ values from the four scattering angles at 30°, 40°, 50°, and 60° are presented. We find two surfaces at which the two temperatures of 60 and 70 °C are almost identical and the Rayleigh ratio is unity, while at 50 °C the Rayleigh ratio is 0.13. For the thermal polymerization of styrene at 60, 70, and 50 °C, no appreciable regular diisopropyl ether was observed. The Rayleigh ratio was computed according to

$$R_{\text{c}}(R) = \frac{I_0 - I_1}{I_0} \quad (14)$$

where I_0 and I_1 are the scattered intensities at 0° and θ , respectively.

C. Thermal Polymerization of Styrene. In order to make simultaneous measurements of Raman scattering and light scattering at the four scattering angles, we have used the same light and the ambient volume corrections of dilute solution of polystyrene (NBS 706 standard) in cyclohexane and an aqueous suspension of 0.06-molar styrene solution. The scattering angles are 30°, 40°, 50°, and 60°. The Rayleigh ratios are given in Table I. Figure 19 shows the Rayleigh ratio as a function of time of polymerization of styrene at 60, 70, and 50 °C, no appreciable regular diisopropyl ether was observed. The Rayleigh ratio was computed according to

where we have neglected the refractive index correction to the initial scattered intensity of the object molecule. Only the mean $\langle r^2 \rangle$ values from the four scattering angles at 30°, 40°, 50°, and 60° are presented. We find two surfaces at which the two temperatures of 60 and 70 °C are almost identical and the Rayleigh ratio is unity, while at 50 °C the Rayleigh ratio is 0.13. For the thermal polymerization of styrene at 60, 70, and 50 °C, no appreciable regular diisopropyl ether was observed. The Rayleigh ratio was computed according to

where I_0 and I_1 are the scattered intensities at 0° and θ , respectively.

The $\langle r^2 \rangle$ values in Figures 17–19 are generally close to the $\langle r^2 \rangle$ and $\langle r^4 \rangle/(\langle r^2 \rangle)^2$ values in Table I.

Table I shows $R_{\text{c}}(R)$ and the $\langle r^2 \rangle/\langle r^2 \rangle_{\text{NBS}}$ values for the various surfaces similar to those experimentally obtained in styrene at 60, 70, and 50 °C, respectively.

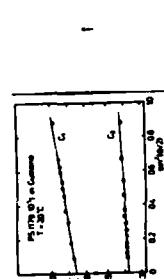
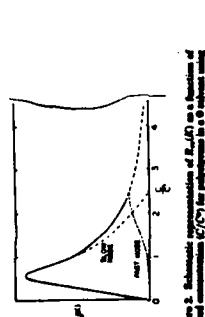
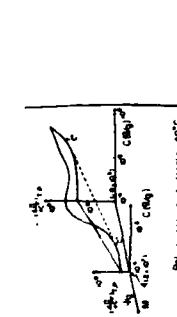
Polystyrene, we found to conduct such surfaces for a more interesting polyphosphine system to be presented in a future article to the present work, let us add to say that the propagation has been discussed. According to Figure 17–19, the Rayleigh ratio of $R_{\text{c}}(70)/R_{\text{c}}(60)$ is 0.13, and the $\langle r^2 \rangle$ and $\langle r^4 \rangle/(\langle r^2 \rangle)^2$ values are nearly the same. The $\langle r^2 \rangle$ and $\langle r^4 \rangle/(\langle r^2 \rangle)^2$ values for the initial polymerization reaction at 60 °C are 1.50 and 0.13, respectively. The Rayleigh ratio of $R_{\text{c}}(70)/R_{\text{c}}(50)$ is 0.13, and the $\langle r^2 \rangle$ and $\langle r^4 \rangle/(\langle r^2 \rangle)^2$ values are nearly the same. The $\langle r^2 \rangle$ and $\langle r^4 \rangle/(\langle r^2 \rangle)^2$ values for the initial polymerization reaction at 50 °C are 1.50 and 0.13, respectively. The Rayleigh ratio of $R_{\text{c}}(60)/R_{\text{c}}(50)$ is 0.13, and the $\langle r^2 \rangle$ and $\langle r^4 \rangle/(\langle r^2 \rangle)^2$ values are nearly the same. The $\langle r^2 \rangle$ and $\langle r^4 \rangle/(\langle r^2 \rangle)^2$ values for the initial polymerization reaction at 60 °C are 1.50 and 0.13, respectively. The Rayleigh ratio of $R_{\text{c}}(70)/R_{\text{c}}(60)$ is 0.13, and the $\langle r^2 \rangle$ and $\langle r^4 \rangle/(\langle r^2 \rangle)^2$ values are nearly the same. The $\langle r^2 \rangle$ and $\langle r^4 \rangle/(\langle r^2 \rangle)^2$ values for the initial polymerization reaction at 60 °C are 1.50 and 0.13, respectively. The Rayleigh ratio of $R_{\text{c}}(70)/R_{\text{c}}(60)$ is 0.13, and the $\langle r^2 \rangle$ and $\langle r^4 \rangle/(\langle r^2 \rangle)^2$ values are nearly the same. 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A study of concentration fluctuations during the thermalization process of dynamic wave-packet correlations was reported by Patterson et al.¹⁷ Here, we extend this analysis to the case of coherent wave-packet correlations with classical wave-packet correlations of thermalizing systems. We assume already that the system has been thermalized. The analysis of the time correlation function of the thermalized system is identical to that of the coherent wave-packet correlation function. The difference is that the thermalized system is characterized by a set of equilibrium parameters, while the coherent wave-packet system is characterized by a set of non-equilibrium parameters. The thermalized system is characterized by a set of equilibrium parameters, while the coherent wave-packet system is characterized by a set of non-equilibrium parameters. The thermalized system is characterized by a set of equilibrium parameters, while the coherent wave-packet system is characterized by a set of non-equilibrium parameters.

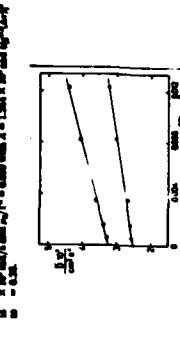
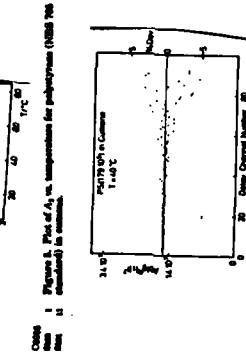
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FIG. 20 (015, 1-2), FIG. 21 (015, 1-2)
FIG. 22 (015, 1-2)

UNIT NO. 127
CAT. NO. MACHINES MARKNOT



UNIT NO. 127
CAT. NO. MACHINES MARKNOT



UNIT NO. 10
DATA SHEET

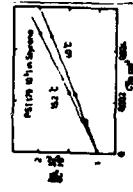


Figure 10. Plot of $\ln(M/M_0)$ vs C_1 for polystyrene in benzene at 25°C.

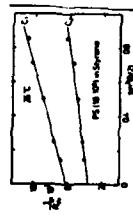


Figure 11. Plot of $\ln(M/M_0)$ vs C_1 for PIB in styrene at 25°C, 50°C, and 80°C.

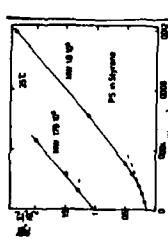


Figure 12. Plot of $\ln(M/M_0)$ vs C_1 for PIB in styrene at 25°C, 50°C, and 80°C.



Figure 13. Plot of $\ln(M/M_0)$ vs C_1 for PIB in styrene at 25°C, 50°C, and 80°C.

UNIT NO. 11
DATA SHEET

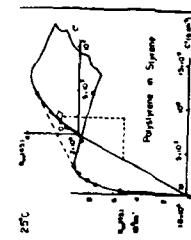


Figure 14. Plot of $\ln(M/M_0)$ vs C_1 for polystyrene in benzene at 25°C.

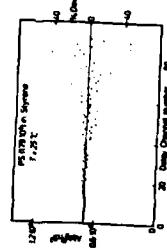


Figure 15. Plot of $\ln(M/M_0)$ vs C_1 for PIB in styrene at 25°C, 50°C, and 80°C.

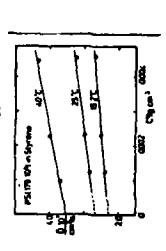


Figure 16. Plot of $\ln(M/M_0)$ vs C_1 for PIB in styrene at 25°C, 50°C, and 80°C.



Figure 17. Plot of $\ln(M/M_0)$ vs C_1 for PIB in styrene at 25°C, 50°C, and 80°C.

Q = 3.62

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ON LINE BURNING MANAGEMENT

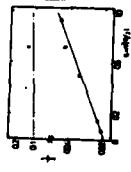


Figure 11. Plot of C_1/C_2 vs C_2/C_3 in the presence of C_3 .

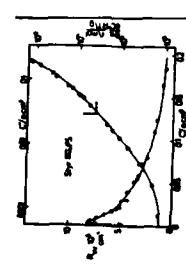


Figure 12. Plot of C_1/C_2 vs C_2/C_3 in the presence of C_1 .

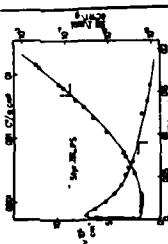


Figure 13. Plot of C_1/C_2 vs C_2/C_3 in the presence of C_1 and C_3 .

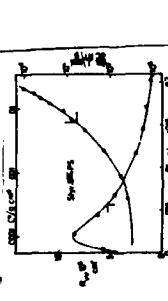


Figure 14. Plot of C_1/C_2 vs C_2/C_3 in the presence of C_1 , C_2 and C_3 .

UNIT NO. 151
ON LINE BURNING MANAGEMENT

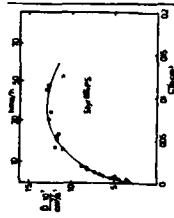


Figure 15. Plot of D_{11}/D_{12} vs D_{12}/D_{13} in the presence of D_{13} .

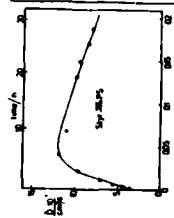


Figure 16. Plot of D_{11}/D_{12} vs D_{12}/D_{13} in the presence of D_{11} .

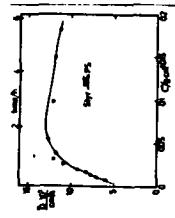


Figure 17. Plot of D_{11}/D_{12} vs D_{12}/D_{13} in the presence of D_{11} and D_{13} .

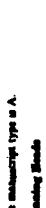


Figure 18. Plot of D_{11}/D_{12} vs D_{12}/D_{13} in the presence of D_{11} , D_{12} and D_{13} .

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Spectroscopic Study of the Phenomenon

Chen and Pyne

Another Author Name

Chu, S. T.-H.

Pyne, G.

Text Paper Size Estimate = 65.7 Page

Graphic Paper Size Estimate = 65.7 Page

Table I
Values of Physical Quantities for Crystalline and Stoichiometric

Table IV
Values of Parameters for Polyisoprene in Système
Polyisoprene A-NBR 70/30 Standard N-1178 Y-161

Volume of Penetrations in $\text{cm}^3 \text{ per } 10^{-2} \text{ cm}^2$		Temperature, °C	
10.0	20	40	60
$A_1 \text{ (from } \text{eqn 1)} \times 10^{-1}$	6.0	6.0	6.0
$A_2 \text{ (from } \text{eqn 1)} \times 10^{-1}$	6.0	6.0	6.0
$M_w \times 10^{-3}$	5.4	5.4	5.4

Reference: B. Polymer Chemical Co., $M_w = 1.3 \times 10^6$

Volume of Penetrations in $\text{cm}^3 \text{ per } 10^{-2} \text{ cm}^2$		Temperature, °C	
10.0	20	40	60
$L_A \times 10^{-1}$	6.0	6.0	6.0
$L_B \times 10^{-1}$	6.0	6.0	6.0
$M_w \times 10^{-3}$	5.4	5.4	5.4

Reference: B. Polymer Chemical Co., $M_w = 1.3 \times 10^6$

Table II
Values of Apparent Characteristic Length L , Second Virial Coefficient A_2 , and Radius of Gyration r_g
in Polyisobutylene (NBS 100 Standard) in Chloroform

Table III Volume of Formation of Polyesters

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